Isotopic Composition of Gadolinium and Neutron-Capture Effects in Some Meteorites[1]

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The isotopic composition of Gd in one chondrite, two achondrites, and the silicate inclusions of two iron meteorites has been determined. When corrected for mass discrimination, Gd in all samples except the Norton County achondrite shows the same relative isotopic abundances as terrestrial Gd. These results set an upper limit of 3×10^{15} neutrons per cm^2 on a differential integrated thermal neutron irradiation of the earth and these meteorites. Neutron-capture effects are present in Gd extracted from the Norton County achondrite. These most probably have been produced by secondary neutrons during the exceptionally long cosmic ray exposure of this large stone meteorite. The isotopic anomalies correspond to an integrated thermal neutron flux of $(6.3 \pm 0.9) \times 10^{15}$ neutrons per cm^2. The percent abundances of terrestrial Gd found in our work for Gd^{160}, Gd^{158}, Gd^{157}, Gd^{156}, Gd^{155}, Gd^{154}, and Gd^{152} are 21.863, 24.835, 15.652, 20.466, 14.800, 2.1809, and 0.2029, respectively. Because of the higher precision, these abundances should replace the currently accepted values.

INTRODUCTION

The large isotopic cross sections of Gd^{157} and Gd^{155} for thermal neutrons (254,000 barns and 60,000 barns, respectively) [*Møller et al.*, 1960] indicate that these isotopes can serve as sensitive indicators of the integrated thermal neutron exposures of natural samples.

In comparing the isotopic composition of Gd in terrestrial samples, meteorites, and lunar material, at least two possible sources for differences have to be considered: (1) a neutron irradiation in the early history of the solar system that was different for terrestrial, meteoritic, and lunar samples and (2) a flux of thermal neutrons produced by moderation of secondary cosmic ray neutrons.

Two factors enter into a general discussion of Gd isotopic variations due to primordial irradiations: (1) the possibility of postirradiation mixing of irradiated and unirradiated material and (2) the magnitude of the integrated neutron flux. At present no positive evidence for such irradiations exists (see *Burnett et al.* [1965, 1966] for detailed discussions). For the case of a very high neutron exposure, e.g., as in the model proposed by *Fowler et al.* [1962] (FGH model), Gd^{157} is totally depleted in the irradiated material; consequently, isotopic variations can result only from differences in the mixing ratios of irradiated and unirradiated material. Previous measurements in four stone meteorites by *Murthy and Schmitt* [1963] showed that meteoritic and terrestrial Gd had the same isotopic composition to within about 1%. For the FGH model, these data allow limits of about 20% to be set for differences in the mixing ratio of irradiated and unirradiated material for terrestrial and meteoritic samples.

Alternatively, it can be assumed that all meteoritic and terrestrial material were subjected to an irradiation while in a highly dispersed state (i.e., a 'uniform irradiation' in the sense of *Burnett et al.* [1966]). In this case an upper limit on the differential thermal neutron exposure of the Gd in terrestrial material and in meteorites can be made. The data of *Murthy and Schmitt* [1963] allow an upper limit of 4×10^{16} neutrons per cm^2 for a differential thermal neutron irradiation to be set.

The neutron-capture effects that we shall describe were recognized by *Daniels* [1953] as an interesting possibility in natural samples.

Because of the great increase in instrumental precision now available [*Wasserburg et al.*,

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1969], and because we have been able to increase the ionization efficiency by about a factor of 50 (see section 2 under Experimental Procedure), it was deemed desirable to reinvestigate this problem. For this purpose, a variety of different meteorite classes were studied. Four criteria were used in selecting the meteorites investigated. (1) In order to study the question of a differential early irradiation of meteoritic and terrestrial matter, the Pasamonte achondrite and the Forest City chondrite, both with a low exposure age (see Table 1), were chosen in an attempt to avoid cosmic ray produced Gd anomalies. (2) The silicate inclusions of the Weekeroo Station and the Copiapo irons were investigated because previous measurements (D. D. Bogard, J. Huneke, D. S. Burnett, and G. J. Wasserburg, unpublished data, 1969) had shown that large concentrations of neutron-capture $Kr^{80, 82}$ and Xe^{128} are present in silicate inclusions from iron meteorites. (3) The Norton County achondrite was chosen on the basis of its high exposure age and its very high recovered weight. The latter fact promises a high slowing down density for secondary spallation-produced neutrons. (4) An attempt was made to sample the principle meteorite classes.

Experimental Procedure

1. Chemical separation. All samples were dissolved in $HClO_4$ and HF, typically 1 ml of each, evaporated to dryness in a stream of filtered nitrogen and redissolved in a few ml of 4*N* HCl. This solution was again evaporated to dryness, the residue dissolved in 2*N* HCl, and the solution stored in a polyethylene bottle.

Gd was separated by ion exchange in a two-column operation. An aliquot corresponding to a 50–100 mg solid sample was taken, evaporated to dryness, and then brought into solution in 2 ml of 1.5 *N* HCl, which was loaded onto a 20 × 1 cm cation-exchange column made from SiO_2 glass, packed with Dowex 50 × 8, 100–200 mesh resin. The resin height after preconditioning with 300 ml 4*N* HCl was 17 cm. Most of the rare earths were separated from all major constituents by elution with 4*N* HCl and collected in the fraction between 40 and 65 ml. This fraction was evaporated to dryness, redissolved in 0.8 *N* HCl and loaded onto the second ion-exchange column.

The purpose of this second column was to separate Gd from the other rare earths. We used a SiO_2 glass capillary with an inner diameter of 0.2 cm packed with carefully sized resin beads of 50–90 μm diameter separated from AG 50W-×4, 200–400 mesh cation-exchange resin.

About 50 ml of the sized resin was typically pretreated as follows: To remove traces of rare earths and Ba, the resin was washed with one liter 4*N* HCl, and the acid was removed with H_2O (five times distilled). Thereupon, the resin was transformed to the NH_4^+ form with one liter of 0.4 *M* 2-methyl lactic acid (E. H. Sargent and Co.) adjusted to *p*H 4.6 with high purity concentrated NH_4OH. Tests with Ce^{144} tracer

TABLE 1. Some General Data on the Investigated Meteorites

Meteorite	Sample Number	Classification[a]	Recovered Weight, kg[a]	Cosmic Ray Exposure Age, m.y.	Gd Concentration, ppm
Pasamonte	297 yII	Eucrite	4	4[b]	2.7[e]
Weekeroo Station	NC-R2	Octahedrite	94	~600[d]	0.3[f]
Copiapo	B2	Octahedrite	40	...	0.2[f]
Norton County	523.3 x Breccia IV	Aubrite	1050	240[c]	0.25[f] 0.38[e]
Forest City	49 t I	Chondrite	122	4.6[b]	0.40[e]

[a] *Hey* [1966].
[b] Based on He^3 concentration [*Kirsten et al.*, 1963] and He^3 production rate of 2×10^{-8} cm³ STP He^3/g m.y.
[c] *Begemann et al.* [1957].
[d] *Bogard et al.* [1968].
[e] *Haskin et al.* [1966].
[f] This work (see section 3 under results).

showed that this *p*H was high enough to prevent resin absorption of rare-earth elements that might have been present in the solution. On the other hand, any Ba present in the solution (see section on interfering ions) would have been strongly held on the resin. Finally, the resin, now in the NH_4^+ form, was washed with H_2O to remove the reagent and the upper third of the resin that might have been contaminated with Ba was discarded.

About 1 cm^3 of the remaining resin was packed into the SiO_2 glass capillary and the resin height adjusted to about 32 cm. Excess water was removed, and 0.2 *M* 2-methyl lactic acid, adjusted to *p*H 4.10 with concentrated NH_4OH was loaded onto the column. The column was allowed to drain for several hours, then a nitrogen pressure of about 0.14 kg/cm^2 over atmosphere was applied to the top of the column for about half an hour to bring the total volume of effluent to 10 ml. Excess solution and excess resin over exactly 30-cm column height were removed with a pipette, and 0.15 ml H_2O was passed through the column. The sample was loaded onto the column after it was prepared to this state. Elution was carried out with 0.2 *M* 2-methyl lactic acid of *p*H 4.10 at a pressure of 0.14 kg/cm^2 over atmosphere resulting in a flow rate of one drop per minute. All separations were carried out at room temperature (22 ± 1°C). After each sample separation, the resin was discarded.

Repeated calibrations of the column with Gd^{155} tracer showed that the peak of the Gd elution curve was reproducible to within an average deviation of ±4 drops for a given batch of resin. However, a shift of 4 drops in the elution curve toward the Eu peak should be easily detected during the mass spectrometric analysis because of the high sensitivity of Eu. Only one column separation out of nine showed serious Eu contamination. The results of this separation were not accepted. Consequently, for reasons that are not understood, it appears that the Gd elution peaks for the samples analyzed were well within the ±4-drop spread. Figure 1 shows that the procedure outlined above resulted in a good separation of Gd from the other rare earth elements.

Blank determinations showed that the Gd contamination introduced by the complete chemical processing was less than 3×10^{-11} gram.

2. *Mass spectrometry.* The sample was evaporated to dryness, redissolved in a drop of 1.5 *N* HCl, and loaded on a rhenium filament. The amount of sample loaded is given in Tables 2–5. For most of the analyses, zone-refined, high-purity Re (The Rembar Company, Inc., Dobbs Ferry, N.Y.) was used after it had been outgassed for two hours at ~2000°C in a vacuum

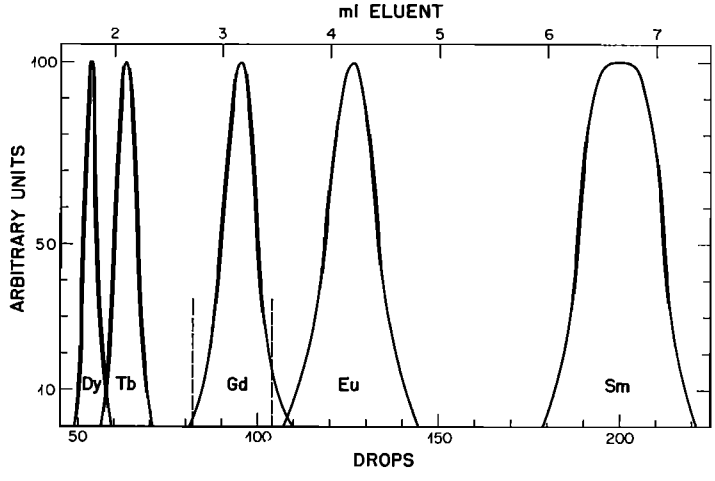

Fig. 1. Chemical separation of Gd from other rare earths by ion exchange. The eluent used was 0.2*M* 2-methyl lactic acid. The calibration was carried out with radioactive tracers of the elements shown. The vertical lines at drops 82 and 104 indicate the fraction of the Gd peak that was collected. It corresponds to about 96% of the total peak.

TABLE 2. Isotopic Ratios of Terrestrial Gadolinium

Run No.[a]	Sample	Sample Size, 10^{-9} gram	Sets of 10 Ratios Taken with Multiplier (MP) or Faraday Cup (FC)	Gd^{158}/Gd^{160}	Gd^{157}/Gd^{160}	Gd^{156}/Gd^{160}[b]	Gd^{155}/Gd^{160}	Gd^{154}/Gd^{160}	Gd^{152}/Gd^{160}
29/30/31 (B)	Reagent Gd	500–2000	10 (FC)	1.1359 ± 0006	0 7155 ±.0006	0.9361 ...	0.6769 ± 0002	0.09971 ± 00006	...
30/31 (B)	Reagent Gd	2000	12 (MP)	1.1353 ±.0005	0 7161 ±.0003	0 9361 ...	0 6767 ± 0004	0.09984 ±.00007	0 00928 ± 00002
51 (A)	Reagent Gd	2000	10 (FC)	1.13590 ± 00009	0.71589 ±.00004	0 9361 ...	0.67695 ±.00009	... ...	... ..
53 (A)	Reagent Gd	2000	12 (FC)	... ...	... ...	0.9361 ...	0.67688 ± 00009	0.09975 ±.00002	... ...
40 (B)	Diabase Wl	50	7 (MP)	1.1357 ± 0007	0.7161 ±.0007	0 9361 ...	0 6771 ±.0003	0 09966 ± 00023	0 00930[c] ±.00004
Adopted value for terrestrial Gd				1.13590 ±.00009	0.71589 ±.00004	0 9361 ...	0 67692 ±.00009	0.09975 ± 00002	0.00928 ± 00002
Collins et al. [1957][d]				1.124 ±.011	0.717 ± 007	0.9361 ...	0.683 ±.007	0.1004 ± 0013	0 00915 ± 00050

[a] (A) and (B) indicate that all sets that were averaged had standard deviations of the ratios $Gd^{155-158}/Gd^{160}$ of less than 0.25 or 0.4%, respectively.
[b] Fractionation correction normalized to $Gd^{156}/Gd^{160} = 0.9361$.
[c] See section 2 under Interfering Ions.
[d] Normalized to $Gd^{156}/Gd^{160} = 0.9361$.

of $\sim 10^{-6}$ torr. After outgassing, no detectable Gd could be found by using the mass spectrometer. After the sample was loaded on the filament, the Gd was transformed to the oxide by heating the filament in air to ~800°C for about one minute. The Lunatic 1 mass spectrometer [*Wasserburg et al.*, 1969] was used for the analyses. GdO ions were obtained by using the single filament surface ionization technique. The stable ion beam was highest for a filament temperature of about 1400°C. The yield for GdO ions obtained by using this technique was about one order of magnitude better than that for Gd ions when a triple Re filament was used. Gd/GdO for the single filament was <0.001, and Gd/GdO for the triple filament was about five. The ratio of GdO ions arriving at the collector to Gd atoms loaded on the filament was about 4×10^{-4} for a sample size of 8×10^{-9} gram Gd. For smaller samples this ratio was even higher. Averages for the isotopic ratios for a set of ten spectra were calculated, cor-

TABLE 3. Isotopic Ratios of Terrestrial Gd after Exposure to Thermal Neutrons

Run No.[a]	Sample	Sample Size, 10^{-9}g	Sets of 10 Ratios taken with Multiplier (MP) or Faraday Cup (FC)	Integrated Neutron Flux (10^{15} Neutrons/cm^2)	Gd^{158}/Gd^{160}	Gd^{157}/Gd^{160}	Gd^{156}/Gd^{160}[b]	Gd^{155}/Gd^{160}	Gd^{154}/Gd^{160}	Gd^{152}/Gd^{160}
56 (A)	GdF_3	~2000	8 (FC)	59	1.14325 ±.00006	0 70628 ± 00009	0 9361 ..	0 67323 ±.00007	0 09949 ± 00003	0 00949[c] ± 00005
58 (A)	GdF_3	~2000	15 (FC)	6.1	1.13664 ± 00016	0.71494 ± 00010	0.9361 ...	0 67649 ± 00012	0 09978 ± 00005	0.00969[c] ± 00005
54 (A)	GdF_3	~2000	12 (FC)	6.1	1.1367 ± 0004	0.7149 ±.0003	0 9361 ...	0.6765 ± 0004	... ...	... .
Terrestrial Gd					1.13590 ±.00009	0.71589 ±.00004	0 9361 ...	0 67692 ± 00009	0.09975 ±.00002	0 00928 ±.00002

[a] (A) and (B) indicate that all sets that were averaged had standard deviations of the ratios $Gd^{155-158}/Gd^{160}$ of less than 0.25 or 0.4%, respectively.
[b] Fractionation correction normalized to $Gd^{156}/Gd^{160} = 0.9361$.
[c] See section 2 under Interfering Ions.

TABLE 4. Gd Isotopic Ratios of Meteorites Showing No Neutron-Capture Effects

Run No.[a]	Sample	Sample Size, 10^{-9}g	Sets of 10 Ratios Taken with Multiplier (MP) or Faraday Cup (FC)	Gd^{158}/Gd^{157}	Gd^{158}/Gd^{160}	Gd^{157}/Gd^{160}	Gd^{156}/Gd^{160}[b]	Gd^{155}/Gd^{160}	Gd^{154}/Gd^{160}	Gd^{152}/Gd^{160}
34 (B)	Pasamonte (separation 1)	10	6 (MP)	1.5874 ±.0013	1.1361 ±.0003	0.7157 ±.0005	0.9361 ...	0.6773 ±.0005	... ...	... ...
37 (B)	Pasamonte (separation 2)	20	8 (MP)	1.5875 ±.0021	1.1360 ±.0009	0.7156 ± 0007	0 9361 ...	0.6766 ±.0005	0 09971 ± 00016	0.00932 ± 00004
55 (B)	Pasamonte (separation 2)	80	4 (FC), 9 (MP)	1.5861 ±.0011	1.1355 ±.0006	0.7159 ±.0003	0 9361 ...	0.6768 ±.0006	0 09971 ±.00005	... ...
39 (B)	Weekeroo Station	11	7 (MP)	1.5868 ±.0022	1.1358 ± 0011	0.7158 ± 0008	0.9361 ...	0.6773 ± 0003	0.09971 ± 00007	0 00938[c] ±.00004
45 (B)	Copiapo	6	6 (MP)	1.5852 ±.0023	1.1358 ±.0007	0 7165 ± 0010	0.9361 ...	0.6772 ± 0005	0.09979 ±.00020	0.00941 ± 00002
61 (B)	Forest City	20	8 (MP)	1.5869 ± 0011	1.1359 ±.0005	0.7158 ±.0004	0 9361 ..	0.6772 ±.0006	0.09976 ±.00005	0.00959[c] ±.00004
Terrestrial Gd				1.5867 ±.0002	1.13590 ±.00009	0.71589 ±.00004	0 9361 ...	0.67692 ±.00009	0 09975 ± 00002	0.00928 ±.00002

[a] (A) and (B) indicate that all sets that were averaged had standard deviations of the ratios $Gd^{158-152}/Gd^{160}$ of less than 0.25 or 0.4%, respectively.

[b] Fractionation correction normalized to $Gd^{156}/Gd^{160} = 0.9361$.

[c] See section 2 under Interfering Ions.

rected for the contribution of O^{17} and O^{18}, and normalized to $Gd^{156}/Gd^{160} = 0.9361$ to correct for mass fractionation. At the beginning of an analysis, this ratio usually was about 0.940 (Faraday cup) and dropped to about 0.935 toward the end of the run. Tables 2–5 give the number of sets of ten spectra obtained for each run. One set of ten spectra including the zero readings on both sides of each peak could be taken within about 15 minutes. The ion beam

TABLE 5. Isotopic Ratios of Gd in the Norton County Achondrite

Run No.[a]	Separation	Sample Size, 10^{-9}g	Sets of 10 Ratios with Multiplier (MP) or Faraday Cup (FC)	Gd^{158}/Gd^{157}	Gd^{158}/Gd^{160}	Gd^{157}/Gd^{160}	Gd^{156}/Gd^{160}[b]	Gd^{155}/Gd^{160}	Gd^{154}/Gd^{160}	Gd^{152}/Gd^{160}
48 (B)	1	12	9 (MP)	1.5907 ±.0019	1.1364 ± 0007	0 7144 ± 0007	0.9361 ...	0.6766 ± 0005	0 09965 ± 00015	0.00944[c] ±.00004
52 (B)	1	12	10 (MP)	1.5914 ±.0014	1.1374 ±.0007	0.7147 ± 0004	0.9361 ...	0.6766 ±.0005	... ...	... ...
59 (B)	2	14	9 (MP)	1.5909 ± 0014	1.1367 ± 0006	0.7145 ±.0005	0.9361 ...	0.6760 ± 0003	0.09969 ± 00011	0.00938[c] ±.00005
60 (A)	2	86	14 (FC)	1.5910 ± 0006	1.13690 ±.00028	0.71456 ±.00020	0 9361 ...	0 67636 ±.00030	... ...	... ...
60 (A)	2	86	28 (MP)	... ...	... ...	... ...	0.9361 ...	... ...	0 09972 ±.00004	0 00933[c] ±.00001
Average for separation 1 (B)				1.5910 ±.0010	1.13690 ±.00057	0.71457 ±.00028	0 9361 ...	0.67659 ±.00043	0 09965 ± 00015	0.00944[c] ± 00004
Average for separation 2 (A)				1 5911 ± 0006	1.13687 ±.00025	0.71454 ±.00019	0.9361 ...	0.67631 ± 00030	0.09971 ± 00004	0 00933[c] ± 00001
Terrestrial Gd				1.5867 ±.0002	1.13590 ± 00009	0.71589 ± 00004	0 9361 ...	0.67692 ±.00009	0.09975 ±.00002	0.00928 ±.00002

[a] (A) and (B) indicate that all sets that were averaged had standard deviations of the ratios $Gd^{158-152}/Gd^{160}$ of less than 0.25 or 0.4%, respectively.

[b] Fractionation correction normalized to $Gd^{156}/Gd^{160} = 0.9361$

[c] See section 2 under Interfering Ions.

was always very stable and no change due to fractionation could be observed within one set of ten spectra.

Samples of less than 20 ng Gd were analyzed using a 17-stage electron multiplier at a gain of 10^4 and a 10^{10}-ohm resistor. An intensity of eight volts for $Gd^{160}O^{16}$ ions could be obtained for several hours for a sample of 10^{-8} gram Gd. Samples of more than 20 ng Gd could be analyzed using a Faraday cup collector with a 10^{11}-ohm resistor. Within one set of ten spectra, the signals of $Gd^{154-160}$ were always recorded using the same amplifier range to eliminate scale-factor corrections.

The linearity of the 10^{11}-ohm resistor was better than 0.02% for the ion currents obtained (see Figure 1 of *Eugster et al.* [1969]). To determine the linearity of the 10^{10}-ohm resistor, we compared the Gd isotopic ratios obtained using this resistor with those using the 10^{11}-ohm resistor. The agreement obtained was better than 0.03%.

Figures 2*a* and 2*b* show typical beam profiles for masses 170 and 171. The distances *AB*

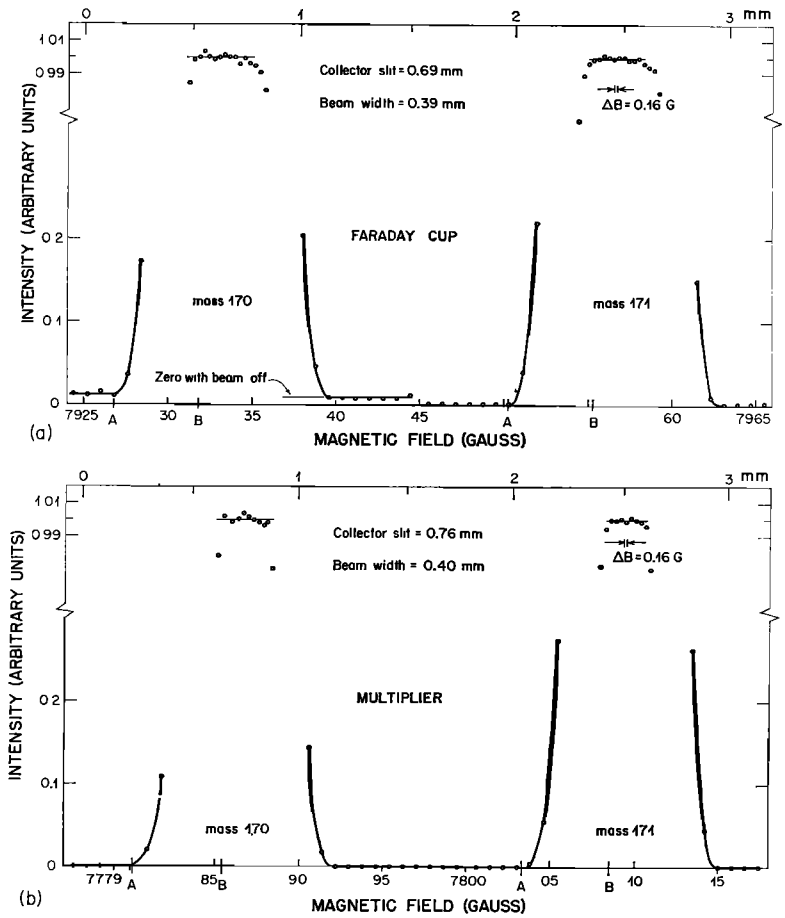

Fig. 2. Beam profiles for Gd at masses 170 and 171 for collector slit wider than beam width and beam intensities typical for meteorite analyses. The source slit was 0.23 mm. Data points correspond to ion-beam currents at appropriate magnetic field values obtained by step scanning. At masses 170 and 171 the integrating times were 8 sec and 2 sec, respectively, for each data point. The gauss equivalent in mm is given on the top axis. a) Beam profile obtained with Faraday cup. The signals of mass 170 and mass 171 correspond to ion currents of 10^{-13} amp and 6×10^{-13} amp, respectively. b) Beam profile obtained with multiplier. The signals of mass 170 and mass 171 correspond to ion currents of 10^{-14} amp and 6×10^{-14} amp, respectively.

correspond to the beam images at the respective collector slits of 0.39 mm (Faraday cup) and 0.40 mm (multiplier). The uncertainty $\Delta B = 0.16\ G$, corresponding to $\Delta B/B = 2 \times 10^{-5}$ [*Wasserburg et al.*, 1969], of the magnetic field for locking in at a preset field value is negligible for a beam width that is about 0.3 mm narrower than the collector slit. At the intensities used for the sample measurements, the mass 170 and mass 171 peak tops are flat to better than 5×10^{-3} and 2×10^{-3}, respectively.

3. *Error calculation.* For each set of ten ratios, mean values of the isotopic ratios Gd^{m}/Gd^{160} and standard deviations for the ratios were calculated. These standard deviations were used to define 'data categories' by means of which a given set of ratios could be accepted or rejected. Categories *B* and *A* data represent sets for which the standard deviations for the ratios $Gd^{155-158}/Gd^{160}$ were better than 0.4% and 0.25%, respectively. Any set of data with standard deviations larger than those of category *B* were rejected. Category *A* data are tabulated in Tables 2–5 whenever possible. The isotopic ratios shown in Tables 2–5 are the averages of the means of all sets in the given category. The errors shown in Tables 2–5 are $2 \times [\Sigma_i \Delta_i^2/n\ (n-1)]^{1/2}$, where n is the number of sets of ten ratios, and where Δ_i is the difference between the mean value of the *ith* set and the value given in the tables. Thus the errors given in this work correspond to 95% confidence limits.

Interfering Ions

Although chemical separations were good, we explicitly investigated possible interferences by BaCl ions and by some rare-earth ions, rare-earth oxide ions and rare-earth chloride ions.

1. *BaCl interference.* Since the sensitivity of BaCl ions for surface ionization is relatively high, even a small amount of Ba in the Gd fraction may cause interference problems with the GdO ions. The Ba in the Gd fraction, determined by isotopic dilution, was 10^{-10} gram Ba or less. Most of the Ba could be baked off by keeping the filament in the mass spectrometer for a few hours at a temperature that was slightly lower than the evaporation and ionization temperature of Gd. At the running temperature used for Gd, the ratio of $BaCl^{35}/Ba$ was about 0.005, and that of $Ba^{138}/Gd^{157}O^{16}$ was usually 0.2–0.001, depending on the original Ba/Gd ratio and on the temperature history of the sample. In order to establish the extent of BaCl interference, we always checked BaCl during the data-taking period. The two possible mass positions for this purpose are mass 169 for $Ba^{134}Cl^{35}$ and mass 175 for $Ba^{138}Cl^{37}$. At mass 175, $Tb^{159}O^{16}$ and Lu^{175} may also appear. Since Tb and Lu are essentially mono-isotopic, it is not possible to exclude the presence of these elements completely. Tb is the neighbor of Gd on the heavier mass side, and a tailing of Tb during the rare earth separation could falsify a determination of BaCl based on this mass position. Therefore, we had to measure $Ba^{134}Cl^{35}$ at mass 169 and to verify the absence of $Eu^{153}O^{16}$ by checking $Eu^{151}O^{16}$ at mass 167. By using mass 169 as a basis for the detection of $Ba^{134}Cl^{35}$, the following problem arises: The abundance of $Ba^{138}Cl^{35}$ is ~30 times higher than that of $Ba^{134}Cl^{35}$, so that an upper limit for a possible peak at mass 169 has to be magnified by a factor of 30 to apply the correction for $Ba^{138}Cl^{35}$ that interferes with the $Gd^{157}O^{16}$. This problem was solved by changing the Ba^{134}/Ba^{138} ratio artificially to become about one by adding highly enriched Ba^{134} to the sample after the chemical separation and to measure this Ba ratio before and after each set of Gd spectra. The amount of enriched Ba^{134} added to the sample was always less than 10^{-10} gram. The ratio of Gd^{m}/Ba^{134} in the Ba^{134} spike was checked by adding enriched Gd^{155} and found to be less than 2×10^{-6} for each Gd isotope *m* and thus could not change the composition of Gd in the sample. In all the runs through run 48, the samples were not spiked with Ba^{134}, and therefore the uncertainty in the abundance of Gd^{157} that is most affected by a BaCl contribution is higher than for the runs performed later. For the runs 34, 37, 39, 45, and 48, the BaCl corrections applied were between 0.04% and 0.125%, and a 100% error for each correction was added quadratically to the statistical errors shown in Tables 2–5. For all other runs the BaCl corrections could be proven to be less than 0.01% and were thus negligible. It may be added that BaF/BaCl was ~10, so that the problem would not have been solved by measuring the Gd ions with which the BaF ions interfere.

2. *Interference by other rare earths.* The

following rare-earth ions, rare-earth oxide ions or rare-earth chloride ions could interfere with GdO ions and were checked repeatedly during each run: Er, Lu, Hf, DyO, and CeCl with the GdO masses 170–176 were less than 0.001% for each isotope, and with mass 168 were less than 0.02%. The presence of Yb ions could not be checked, the isotopes of this element occupying exactly the same mass positions as GdO[16]. However, the chemical separation of Yb from Gd was even better than that of Dy and Er, for which no traces were found in the samples, so that the abundance of Yb in the sample must have been extremely low. A contamination of Gd by 0.1% of Yb relative to Gd would increase the ratio Gd^{157}/Gd^{160} by ~0.01% and that of Gd^{158}/Gd^{160} by ~0.05% if the isotopic ratios are normalized to a constant Gd^{156}/Gd^{160} ratio. Hence anomalies resembling those for neutron capture cannot be introduced by an Yb contribution. The same is true for an addition of LaCl to masses 174 and 176 and CeO_2 to masses 172 and 174. Besides, the presence of LaCl and CeO_2 ions was unlikely since La and Ce ions were not detected. LaO_2 would interfere with $Gd^{155}O$ and PrO_2 with $Gd^{157}O$. However, we never observed a systematic variation of the ratios $Gd^{155-158}/Gd^{160}$ although we changed the GdO ion current during the course of the analyses by a factor of up to 100 corresponding to a variation of the filament temperature of about 200°C. We may therefore assume that the contribution of the compounds of the other rare earths to the masses of $Gd^{155-160}O$ that will be used for the discussion of neutron capture effects was negligibly small.

For most of the samples, we observed a systematic enhancement of the abundances of masses 168 ($Gd^{152}O$) and 170 ($Gd^{154}O$) indicating that an Sm contribution was present, caused by the blank of the second ion-exchange column, not by incomplete chemical separation. Although both Gd^{154}/Gd^{160} and Gd^{152}/Gd^{160} are of no significance for the discussion of neutron-capture effects (see section 2 under Discussion), we corrected for Sm in order to ensure that the enrichments of Gd^{154} and Gd^{152} are compatible with a Sm contribution. Sm isotopes that did not interfere with these two Gd isotopes were detectable at the beginning of the runs. However, at the temperature at which data were taken, the ratios Sm/Gd^{160} dropped below 2×10^{-5}. Because of an enhanced background due to deflected ions in the region of the lower SmO masses these small peaks became undetectable. We therefore corrected the measured ratio $(154/160)_{meas}$ based on the measured ratio $(152/160)_{meas}$. Equation 1 pays regard to the fact that for samples with neutron-capture effects Gd^{152} is enriched by neutron capture of Eu^{151}. Furthermore, a mass-fractionation correction for $(152/160)_{meas}$ has to be applied for samples with neutron-capture effects because the mass fractionation calculated by using a constant ratio $(Gd^{156}/Gd^{160}) = 0.9361$ is not the true mass fractionation. The ratio Gd^{154}/Gd^{160} corrected for the Sm contribution was calculated according to equation 1:

$$\begin{aligned}(Gd^{154}/Gd^{160})_{corr} &= (154/160)_{meas} \\ &- \{(152/160)_{meas} \\ &\times [1 + 2(Gd^{155}/Gd^{156})_{terr}(\varphi t)\sigma^{155}] \\ &- (Gd^{152}/Gd^{160})_{terr} \\ &- (Eu^{151}/Gd^{160})_{sample}(\varphi t)\sigma^{155}\} \\ &\times (Sm^{154}/Sm^{152})_{terr} \qquad (1)\end{aligned}$$

Here the terrestrial Gd isotopic ratios are as given in Table 2 and $(Sm^{154}/Sm^{152})_{terr} = 0.845$ [*Collins et al.*, 1957]. (φt) is the neutron flux in the sample calculated using Equation 9 and $\sigma^{155} = 62{,}500$ barns (see section 2 under Discussion). For σ^{151}, the thermal neutron-capture cross section of Eu^{151} for the production of Gd^{152}, we used the value of 4000 barns.

The Sm correction of all analyses of reagent Gd was less than 0.01% for Gd^{154} and less than 0.1% for Gd^{152}, and thus negligible. This could be established because relatively high ion beams were obtained for the relatively big amounts of reagent sample loaded on the filament.

The ratios Gd^{152}/Gd^{160} given in Tables 2–5 are always the values uncorrected for any possible interferences, whereas those of Gd^{154}/Gd^{160} are always corrected for Sm^{154} based on the measured Gd^{152}/Gd^{160} using equation 1. The Sm correction to Gd^{154} was always less than 0.3%. The isotopes $Gd^{155-160}$, which are important for neutron capture effects, are not affected by a possible Sm contribution.

3. *Hydrocarbons.* By closing the collector slit from the regular width (see Figures 2*a* and

2*b*) to 0.25 mm, hydrocarbon peaks could be partially resolved from the GdO peaks. No hydrocarbons (less than 0.01% for each Gd isotope) could be detected in the GdO region at the time that measurements were being made.

4. *Oxygen correction.* In all runs GdO ions were measured. In order to obtain the isotopic ratios of the Gd ions, small corrections for the compounds of the two neighboring Gd isotopes on the lower mass side with O^{17} and O^{18} have to be applied. As described earlier, the averages of the ratios of the ion beam readings at the masses where GdO^{16} occurred (168/176, 170/176, 171/176, 172/176, 173/176 and 174/176) were calculated for a set of ten spectra. Thereupon, corrections for the contributions of GdO^{17} and GdO^{18} to the above masses were calculated according to equations 2–7:

$$(168/176)_{corr} = 1.00232 \times (168/176)_{meas} \quad (2)$$

$$(170/176)_{corr} = 1.00213 \times (170/176)_{meas} \quad (3)$$

$$(171/176)_{corr} = 1.00226 \times (171/176)_{meas} \quad (4)$$

$$(172/176)_{corr} = 1.00183 \times (172/176)_{meas} \quad (5)$$

$$(173/176)_{corr} = 0.99990 \times (173/176)_{meas} \quad (6)$$

$$(174/176)_{corr} = 1.00040 \times (174/176)_{meas} \quad (7)$$

Here the ratios indicated by the subscript 'meas' are the measured ratios (uncorrected for fractionation), whereas those indicated by 'corr' are the ratios corrected for the contributions of the compounds of Gd with O^{17} and O^{18}. Equations 2–7 yield the ratios of the Gd isotopes uncorrected for mass discrimination. All data presented in this work relate to Gd after correction for O^{17} and O^{18}.

For the ratios O^{18}/O^{16} and O^{17}/O^{16}, we used the values as given by *Nier* [1950], who quotes errors of 0.1% and 0.7%, respectively. In addition to these statistical errors, there might be systematic errors of the oxygen isotopic ratios caused by a mass fractionation that we estimate to be less than 1% per mass unit. Therefore we assume the values used for the isotopic ratios of oxygen to be correct within ±2%. Using values of the isotopic ratios O^{18}/O^{16} and O^{17}/O^{16} differing by 2%, the correction factors in equations 2–7 change by less than five parts in 10^5, which is negligible compared to our experimental uncertainty.

Furthermore, any systematic errors in the oxygen corrections would not affect a comparison between different samples with small anomalies.

After the Gd isotopic ratios of a set of ten spectra were corrected as outlined above, the isotopic ratios were then corrected for mass fractionation by normalization to $Gd^{156}/Gd^{160} = 0.9361$. This value corresponds to the typical (to within 0.5%) ratios Gd^{156}/Gd^{160} (not corrected for mass fractionation), which were measured on reagent Gd using the Faraday cup. It was chosen as basis of the mass fractionation correction in order to keep the correction as small as possible. This is distinct from the literature value of *Collins et al.* [1957].

Results

1. *Terrestrial gadolinium.* In Table 2 the isotopic ratios obtained for terrestrial Gd are compiled. The contributions of BaCl and Sm were less than 0.01% for all runs in Table 2. The mass fractionation correction was made by normalizing the $Gd^{156}/Gd^{160} = 0.9361$. The different measurements agree within the statisitical errors, and no systematic differences were found between data obtained using the multiplier (MP) and those obtained using the Faraday cup (FC).

The excellent reproducibility of the analyses tabulated in Table 2 shows that the calculated statistical error provides a realistic estimate of the over-all precision for a given mass spectrometric analysis. Further, these indicate that isotopic variations of 0.1% in the $Gd^{155-156}/Gd^{160}$ should be detected even for small (less than 10^{-7}-gram samples).

Table 2 also gives values for the terrestrial Gd isotopic ratios as determined by *Collins et al.* [1957]. Our results agree reasonably well with the data to within the precision that they report. Although there undoubtedly is a small systematic error due to mass discrimination in both the present and the literature data, it is reasonable to use the abundance data reported in the present work for Gd over that of previous workers because of the higher precision of the new data. The per cent abundances of terrestrial Gd found in our work for Gd^{160}, Gd^{158}, Gd^{157}, Gd^{156}, Gd^{155}, Gd^{154}, and Gd^{152} are 21.863 ± 0.002, 24.835 ± 0.004, 15.652 ± 0.002, 20.466 ± 0.002, 14.800 ± 0.003, 2.1809 ± 0.0006, 0.2029

± 0.0005, respectively. It should be emphasized that these values are *relative* abundances. The errors given do not include the uncertainty due to the unknown mass fractionation.

2. *Irradiation experiment.* We have analyzed irradiated Gd samples in order to confirm the expected sensitivity for detecting small neutron-capture effects of Gd^{157} and Gd^{155}. Thin layers of GdF_3 were vaporized in vacuum onto high-purity Al foils. To prevent self-shielding effects the thickness was adjusted to about 10^{-4} gram GdF_3/cm^2. Two samples were exposed to a well-thermalized neutron flux in the Brookhaven Medical Reactor together with two Au monitors located above and below the Gd sample. One exposure was 10 min, the other was 100 min. A sample of Cd-covered Au was exposed separately to correct for epithermal contributions to the observed Au^{198} activity. After the irradiation the thermal neutron fluxes were determined, based on the Au^{198} γ activity of the monitors and are given in Table 3. The Gd was removed from the Al foil by washing off with ~0.5 N HCl. The two flux monitors within each sample gave the same flux to within 5%. The measured integrated fluxes for the irradiations agreed with the flux values given by the reactor operators to better than 10%.

The results obtained are shown in Table 3. All data are normalized to the same ratio $Gd^{158}/Gd^{160} = 0.9361$ as the terrestrial samples (i.e., an effective discrimination is calculated assuming this ratio). A normalization procedure based on Gd^{154}/Gd^{160} is more suited for the discussion of neutron-capture anomalies since Gd^{154} and Gd^{160} are not affected. However, the precision of this ratio is in most cases considerably lower than for $Gd^{155-158}/Gd^{160}$. We have made allowance for the fact that the discrimination calculated by using $Gd^{156}/Gd^{160} = 0.9361$ is not the true discrimination in samples that have been changed by neutron capture.

This can be taken into consideration in a plot of Gd^{158}/Gd^{160} versus Gd^{157}/Gd^{160} (both normalized to $Gd^{156}/Gd^{160} = 0.9361$). To first order, the correlation curve of these ratios is a line with the slope

$$\frac{d(158/160)_{norm}}{d(157/160)_{norm}} = -\frac{[1 - 0.5(\sigma^{155}/\sigma^{157})(155/156)_{terr}(158/157)_{terr}]}{[1 + 0.75(\sigma^{155}/\sigma^{157})(155/156)_{terr}]} = -0.764 \qquad (8)$$

For $\sigma^{155}/\sigma^{157}$ we used the value of 0.2386 [*Møller et al.*, 1960].

Figure 3 shows the correlation line calculated according to equation 8 and the data points for the irradiated samples. For changes by neutron capture of Gd^{157}/Gd^{160} of 2% or less, the slope of the correlation line changes by less than 0.13%. The experimental data agree exactly with the calculated line. If we had normalized the Gd isotopic ratios to a constant ratio Gd^{154}/Gd^{160}, the slope of the corresponding correlation line would be −1.

The cross-section ratio and hence the correlation line is very insensitive to neutron energy in the thermal region.

Note that the slope of the correlation line depends only on $\sigma^{155}/\sigma^{157}$ and not on the integrated neutron flux. The integrated flux determines the position of a given point along the correlation line.

Figure 4 shows the correlation of Gd^{155}/Gd^{160} versus Gd^{157}/Gd^{160}, normalized to $Gd^{158}/Gd^{160} = 0.9361$. The correlation line has a slope of 0.3797 that changes by less than 0.3% for variations of the ratio Gd^{157}/Gd^{160} of 2% or less.

Although the capture cross section is not strictly proportional to $1/V$, neutron capture of Gd^{157} can be described by an average value of $\sigma V = 4.75 \times 10^{-14}$ cm/sec (see section 1 under Discussion). Using this and the thermal neutron density calculated from the measured Au^{198} activity, the expected percentage changes in the corrected Gd^{158}/Gd^{157} ratios would be 2.1% and 0.22% for the long and short irradiations, respectively. The corresponding measured changes of (2.02 ± 0.03)% and (0.20 ± 0.03)% are in excellent agreement with the above predictions.

The analyses of the irradiated samples have thus demonstrated that 0.1% isotopic variations due to neutron capture can be readily detected. This confirms the sensitivity predicted from the precision of the measurements on terrestrial samples. The irradiation experiments also indicate that reliable neutron fluxes can be calculated from Gd isotopic measurements using

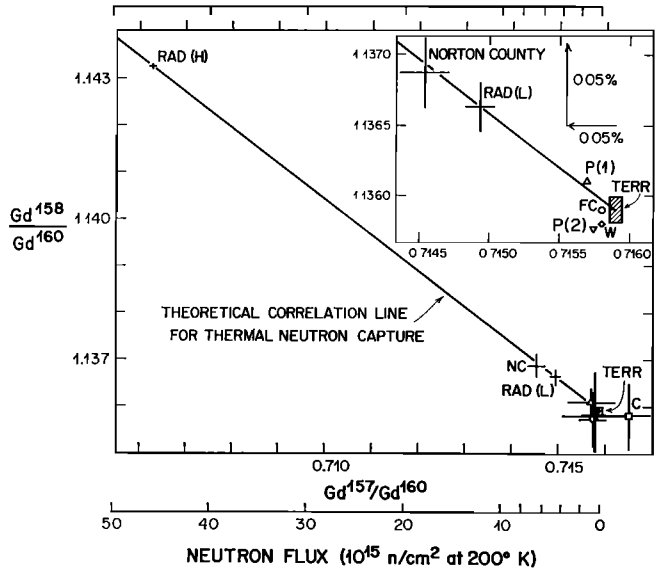

Fig. 3. Gd^{158}/Gd^{160} versus Gd^{157}/Gd^{160} correlation diagram. Changes of the isotopic ratios of 0.05% are indicated by the two arrows shown. The scale for the neutron flux corresponding to the Gd^{157}/Gd^{160} values is calculated for σ^{157} (200°K) = 262,000 barns. For σ^{157} (293°K) = 215,600 barns and for σ^{157} (373°K) = 179,200 barns, the scale for the neutron flux has to be multiplied by 1.22 and 1.46, respectively. P(1), Pasamonte separation 1; P(2), Pasamonte separation 2; W, Weekeroo Station; C, Copiapo; FC, Forest City; NC, Norton County; RAD(H), highly irradiated terrestrial Gd; RAD(L), low-irradiated terrestrial Gd; TERR, terrestrial Gd. The squares, triangles, etc. represent corresponding points on the main and inset figure. For better clarity, the error bars for the meteorites showing no neutron-capture effects and the point for Copiapo are not shown in the inset figure.

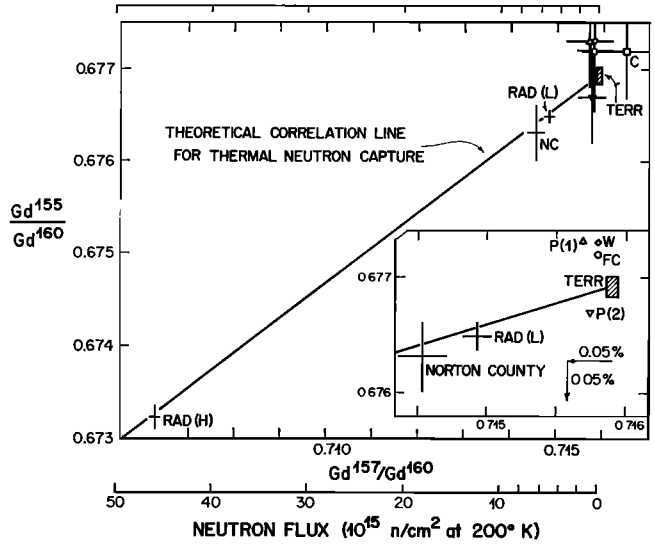

Fig. 4. Gd^{155}/Gd^{160} versus Gd^{157}/Gd^{160} correlation diagram. See caption of Figure 3.

the available laboratory neutron cross section data.

3. *Gd concentrations.* For samples for which the Gd concentrations were not known, a small aliquot of the Gd sample was spiked after the chemical separation with enriched Gd^{155} and analyzed. Since the Gd yield of the first ion-exchange column used was more than 99% and that of the second one about 96% (see Figure 1), we estimate our Gd concentrations given in Table 1 to be correct within about ten per cent.

4. *Meteorites showing no Gd anomalies.* Table 4 gives the data for meteorites showing no neutron capture anomalies. Within the limits of experimental errors, the results for each run agree with the results for terrestrial Gd. The quality of the data differs considerably for different runs, depending mainly on the size of the sample analyzed. A small BaCl correction had to be applied for the runs of very small samples (runs 34, 37, 39, and 45) and a 100% error for each correction was added quadratically to the statistical errors. Therefore, these data are of somewhat lower quality than those for which the BaCl correction was negligibly small.

For Pasamonte, two different chemical separations and three different mass-spectrometer runs of widely varying sample sizes were performed. The reproducibilities of the ratios $Gd^{155-158}/Gd^{160}$ for the different chemical separations and also for the different runs of the same chemical separation are well within the experimental errors of less than 0.1%. The most precise analysis of Pasamonte (run 55) is about of the same quality as that of Forest City. For both runs the BaCl correction was negligible, and the precision is about 0.05% for the ratios of the abundant Gd isotopes.

The samples of the silicate inclusions of the two iron meteorites, Copiapo and Weekeroo Station, were very small. The Gd isotopic composition of Weekeroo Station agrees well with that of terrestrial Gd although the error limits are about 0.1%. Copiapo, for which a sample of only 6×10^{-9} gram Gd was analyzed and for which the highest BaCl correction had to be applied, shows the least precise value of the ratio Gd^{157}/Gd^{160}. However, since no neutron-capture anomalies are evident within experimental errors, we considered it unnecessary to repeat the analysis. The data points of the meteorites showing no neutron-capture anomalies are shown in Figures 3 and 4. The ratios Gd^{157}/Gd^{160} for Pasamonte, Weekeroo Station, and Forest City lie within 0.04% of the data point for terrestrial Gd.

5. *The Norton County achondrite.* Table 5 gives the results for the Norton County achondrite. We carried out two different chemical separations. For separation 2, we processed four aliquots of 100-mg solid sample through the first ion-exchange column. Then we combined the four rare-earth fractions for the separation of Gd using the second ion-exchange column. For each separation, two mass-spectrometer runs were performed. Inspection of the data shows that for each run significant anomalies are present compared to terrestrial Gd. The results for all four runs agree within the experimental errors. For run 48, a BaCl correction of 0.1% and less had to be applied; for all other runs, the BaCl contribution was negligibly small. Samples of $\sim 13 \times 10^{-9}$ gram Gd were analyzed using the multiplier with statistical error limits of about 0.07% for the ratios $Gd^{155-158}/Gd^{160}$. The data for the sample of 86×10^{-9} gram Gd were obtained using the Faraday cup, and the experimental errors are two to three times smaller than those of the three other runs. The data shown for the average of separation 2 were obtained by calculating average values for those data sets from runs 59 and 60 whose standard deviations were less than 0.25% for the ratios $Gd^{155-158}/Gd^{160}$ and less than 0.5% for the ratio Gd^{154}/Gd^{160}. The average values for separation 1 agree almost exactly with those for separation 2. The more precise data are shown in Figures 3 and 4. The agreement with the neutron-capture correlation lines is very good, and we can conclude that the anomalies found in Norton County are caused by a thermal neutron flux.

Discussion

1. *Irradiation models.* As indicated in the Introduction, the conclusions drawn from the Gd isotopic measurements depend on the irradiation model adopted. We discuss three simple types of irradiations:

a. In-situ irradiation of the meteorites at some time subsequent to their formation, e.g., by galactic cosmic rays or solar flare neutrons.

b. A uniform primordial irradiation of both

meteoritic and terrestrial matter at a time when they were sufficiently dispersed so that all the material was irradiated. For both *a* and *b* identical values for integrated neutron fluxes are calculated. However, for *b* the calculated flux is the *difference* in the integrated fluxes experienced by terrestrial and meteoritic material, whereas for *a* the calculated value is the *magnitude* of the flux because we assume that terrestrial material has not been irradiated.

c. An intense primordial irradiation of only a fraction of the solar system material followed by mixing of the irradiated and unirradiated fractions (e.g., as suggested by *Fowler et al.* [1962]). Gd^{157} and Gd^{155} are assumed to be totally depleted in the irradiated material. In this case, Gd isotopic variations reflect variations in the mixing ratio of irradiated and unirradiated material rather than flux variations. Models *b* and *c* are extremes. A continuum of intermediate models exists.

2. *Effective capture cross sections of Gd.* The large thermal cross sections of Gd^{155} and Gd^{157} are due to low-energy resonances; however, the resonance energies (0.0268 and 0.0314 ev, respectively) are sufficiently low that the $1/V$ behavior dominates the energy dependence of the cross section. Thus, below about 0.04 ev, the cross sections are approximately $1/V$. We assume that the neutrons with energies below about 0.1 ev undergo a sufficient number of collisions to attain a Maxwell-Boltzmann energy distribution. Thus, because 0.04 ev is higher than the kT values of interest, the value of σV averaged over a thermal distribution $\langle \sigma V \rangle$ should not depend strongly on the temperature. Calculations of $\langle \sigma V \rangle$ using the single level Breit-Wigner resonance parameters of *Møller et al.* [1960] showed that $\langle \sigma V \rangle = 4.75 \times 10^{-14}$ cm^3/sec to within $\pm 5\%$ between 100°K and 393°K for Gd^{157}. However, this value is about 20% lower than σV evaluated at $V = 2200$ m/sec, reflecting the fact that the cross section is not strictly proportional to $1/V$. The ratio of $\langle \sigma^{157} V \rangle$ to $\langle \sigma^{155} V \rangle$ is also approximately constant at 4.1 ± 0.1 over this temperature range. Because of the approximate constancy of $\langle \sigma V \rangle$, the neutron density can be determined relatively unambiguously by measurement of Gd^{157} isotopic variations, whereas the calculation of an integrated thermal neutron flux requires the assumption of a temperature. However, deviations of the actual energy spectrum from a Maxwell-Boltzmann distribution are possible for near-surface samples and for samples that are comparatively strong neutron absorbers. We have thus assumed a temperature of 200°K and calculated fluxes and flux limits. The corresponding effective capture cross section for Gd^{157} of 262 kb will be used in all the following calculations.

The isotopic ratio Gd^{158}/Gd^{157} is more sensitive to a neutron flux than are the other ratios measured. We therefore included these ratios in Tables 4 and 5 and calculated the experimental errors by adding quadratically the errors of the measured Gd^{157}/Gd^{160} and Gd^{158}/Gd^{160}. To the first order the integrated neutron fluxes can then be calculated

$$(\phi t) \cong \frac{(Gd^{158}/Gd^{157})_{meas} - (Gd^{158}/Gd^{157})_{terr}}{\sigma^{157}[1 + (Gd^{158}/Gd^{157})_{terr}] + \frac{1}{4}\sigma^{155}(Gd^{158}/Gd^{157})_{terr}(Gd^{155}/Gd^{156})_{terr}}$$

$$= 1.44 \times 10^{18}[(Gd^{158}/Gd^{157})_{meas} - (Gd^{158}/Gd^{157})_{terr}]n/cm^2 \quad (9)$$

The second term in the denominator stems from the normalization of the measurements to $Gd^{158}/Gd^{160} = 0.9361$. This first-order approximation introduces an error of less than 1% in the calculated flux for variations in Gd^{158}/Gd^{157} of 1.3% or less.

3. *Neutron fluxes due to in-situ irradiations.* The data in Table 4 show that any differences in the Gd^{158}/Gd^{157} ratio are less than about 0.002 which, from equation 9, implies an upper limit on the in-situ integrated thermal neutron flux of 3×10^{15} neutrons/cm^2. Using the exposure ages tabulated in Table 1 for Pasamonte and Forest City, this corresponds to a flux of less than about 20 neutrons/cm^2 sec.

This limit is compatible with the cosmic ray thermal neutron fluxes of less than roughly 0.1 neutron/cm^2 sec, which can be estimated from the calculations of *Eberhardt et al.* [1963] for small meteoroids (radius $\leqq$ 25 cm) of chondritic composition.

The Norton County aubrite is a stone remarkable in a number of respects (cf. Table 1): (1) It is the largest stone meteorite existing as a single piece [*Hey*, 1966]. (2) It has the highest cosmic ray exposure age of all stone meteorites by about a factor of four. (3) Norton County was the first stone meteorite for which an exposure age was determined [*Begemann et al.*, 1957]. Today it is interesting to note that these workers were concerned about the *brevity* of the H^3–He^3 exposure age they found compared to the K^{40}–Ar^{40} age of 4.4 b.y. for this meteorite. (4) The iron content of this meteorite is very low (~1.6% total Fe [*Wiik*, 1956]).

For the study of effects caused by thermal neutrons in meteoritic material, the properties (1), (2), and (4) are essential. A high slowing-down density is necessary for moderating the Mev secondary neutrons down to thermal energies, and a high exposure age results in a large neutron dosage. Furthermore, neutron capture by Fe is low.

Table 5 shows that the ratio Gd^{158}/Gd^{157} of Norton County is $(0.27 \pm 0.04)\%$ higher than that of terrestrial Gd. This anomaly corresponds to an integrated neutron flux of $(6.3 \pm 0.9) \times 10^{15}$ neutrons/cm^2.

Using 600 m.y. as an exposure age for Weekeroo Station [*Bogard et al.*, 1968], an upper limit of 0.2 neutrons/cm^2 sec is obtained. In view of this comparatively low limit, the presence of large quantities of $Kr^{80 \text{ and } 82}$ and Xe^{129} due to epithermal neutron capture on $Br^{79 \text{ and } 80}$ and I^{127}, respectively, (D. D. Bogard, J. Huneke, D. S. Burnett, and G. L. Wasserburg, unpublished data, 1969) must be ascribed to a combination of (1) the poor moderating properties of Fe and Ni and (2) comparatively large halogen contents for silicate inclusions in iron meteorites.

The experimental evidence is very strong that the neutron effects found in Norton County are caused by cosmic radiation. *Burnett et al.* [1966] showed that the enrichment of K^{40} in this meteorite is due to the $Ca^{40}(n, p)$ reaction. The amounts present were smaller than those predicted from the cosmic ray spectrum of *Arnold et al.* [1961]. Burnett et al. found a K^{40} production rate for $Ca^{40}(n, p)$ of about 6×10^{13} atoms K^{40}/g Ca. For a cross section for the $Ca^{40}/(n, p)$ reaction of 0.4 barn, this production rate corresponds to an integrated flux of 7.5×10^{15} neutrons/cm^2. Thus, the ratio of the thermal to Mev neutron flux in Norton County is about 1. A similar calculation for Weekeroo Station yields a thermal to Mev flux ratio of less than 10^{-2}. This is presumed to be a consequence of the larger size and superior moderating properties of Norton County.

It is possible to obtain an estimate of the epithermal neutron flux in Norton County from rare-gas data by using an indirect approach. The spallation ratio Ne^{21}/He^3 depends on the hardness of the energy spectrum of the cosmic radiation [*Eberhardt et al.*, 1966]. Neutrons with energies between ten and a few hundred Mev will induce spallation reactions resulting in small ΔA. *Eugster et al.* [1969] have shown that the amounts of neutron-produced Kr^{80} normalized to 1 ppm Br and 10^{-8} cm^3 STP $He_{spall}{}^3$/g in ten chondrites are correlated with the spallation ratio Ne^{21}/He^3. The spallation ratio Ne^{21}/He^3 is thus closely related to the epithermal neutron flux F by the empirical equation

$$F = \frac{s}{\sigma^{79}} [(Ne^{21}/He^3)_{spall} - 0.15] \qquad (10)$$

The value of s is derived from Figure 10 of *Eugster et al.* [1969]. We used 110 barns for σ^{79} [*Marti et al.*, 1966] for the production of Kr^{80} by epithermal (30–300 ev) neutron-capture of Br^{79} and a He^3 production rate of 2×10^{-8} cm^3 STP/g m.y. *Eberhardt et al.* [1965] pointed out that for the same irradiation spectrum Ne^{21}/He^3, ratios are 20% higher in aubrites than in chondrites, so that the measured spallation ratio Ne^{21}/He^3 for Norton County at 0.26 corresponds to a chondritic value of 0.21. Assuming that s is the same for Norton County and for chondrites, this ratio corresponds to a flux of 3×10^{15} neutrons/cm^2 m.y. which, with an exposure age of 240 m.y. for Norton County gives an integrated flux of 6×10^{15} neutrons/cm^2. Because s should be somewhat higher for Norton County, this value may be somewhat low. Nevertheless, it appears that the neutron fluxes in the three energy ranges considered (thermal, epithermal, and fast) are of the same order of magnitude.

Eberhardt, Geiss, and Lutz [1963] (EGL) have calculated neutron fluxes produced by cosmic rays as a function of position and radius for spherical meteoroids of chondritic composi-

tion. Because of the low concentrations of heavy elements, particularly Fe, Norton County is an excellent moderator. The estimated resonance escape probability for Norton County is about 0.7, whereas for chondrites it is 0.1. This means that the approximations in the EGL calculations are better for Norton County than for chondrites. We assume that the pre-atmospheric radius of Norton County was 50 cm, i.e., approximately the same as its terrestrial size, and that our samples were taken near the present surface at $r = 40$ cm. (Deep interior samples of Norton County have never been distributed.) Using the 'analytical method' of EGL and making the necessary changes in the parameters to account for the differences from chondritic composition, a flux of 2 neutrons/cm^2 sec is calculated. This is in good agreement with the measured value of 1.0 neutrons/cm^2 sec. Thus, the EGL calculations and a cosmogenic origin for the Gd isotopic variations in Norton County are mutually consistent. If we consider the cosmogenic origin to be established independently, and if we accept the assumption of similar pre-atmospheric and terrestrial sizes for Norton County, the present experiments provide the first conclusive test of the EGL calculations.

4. *Limits on primordial irradiations.* For a uniform primordial irradiation (Model *c* in section 1 under Discussion), limits on the difference in the fraction q of material irradiated can be set. Following the discussion given by *Burnett et al.* [1966], we obtain a value of q equal or less than 0.001. For example, for a model with 10% of the solar system material irradiated, this limit would mean that the earth and the meteorites investigated (except Norton County) contain the same proportions of irradiated and unirradiated material to within 1%. These limits are over an order of magnitude lower than those previously set.

Conclusions

Gd in the Norton County achondrite shows cosmic ray effects induced by the capture of spallation-produced thermal neutrons by Gd^{157} and Gd^{155}. The integrated thermal neutron flux corresponding to the anomalies found is $(6.3 \pm 0.9) \times 10^{15}$ neutrons/cm^2. The integrated fluxes of epithermal and fast neutrons found based on rare gas data and on K^{40}, respectively, are of the same order of magnitude. The possibility of detecting neutron anomalies of 0.1% in samples of about 10^{-8} gram Gd provides us with an important technique for the study of the history of the lunar surface.

All other iron and stone meteorites investigated in this work contain Gd with the same isotopic composition as terrestrial Gd. This places an upper limit of 3×10^{15} neutrons/cm^2 for a differential primordial thermal neutron irradiation of the earth and the meteorites.

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